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13. ABSTRACT (Maximum 200 words) Studies were carried out in three specific areas of food science and technology. This research was directed toward gaining a real understanding of water in food systems, incorporating the view of water as a reactant and a "catalyst", water as a structure influencing material, and water mobility as a function of external and internal influencing factors. It also was focused on the conversion of extrusion-cooking technology into extrusion-cooking science, considering the mechanical aspects of the extruder as well as the chemistry and kinetics of the extruder as a pressure-shear-temperature reactor. Another component of the studies was dedicated to the development of new in-line sensor concepts for real-time measurement encountered in a variety of food processing environments, through the synthesis of new chemical compounds and new materials which may serve as sensors and the development of new systems which provide measurable responses leading to improved process efficiency and product quality through automation and control.					
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The Center For Advanced Food Technology

Food Related Studies

Final Report

Prepared by:

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November 16, 1992

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Research Agreement Number: DAAL03-89-K-0174

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1. Forward

The research effort carried out in this project leveraged ARO funds with funds from a group of industrial firms and from the state of New Jersey to provide a total investment in excess of \$1.0 million annually. The ARO funds were key to providing the U.S. Army Natick Research, Development and Engineering Laboratories access to the results of the research. The research approach was to study phenomena in well characterized models, gain understanding of underlying mechanisms rather than macro consequences and translate the findings to more complex systems and ultimately to foods.

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3. List of Appendixes, Illustrations and Tables

NONE

4. Body of Report

4.A. Statement of the Problem Studied

Studies were carried out in three specific areas of food science and technology.

This research was directed toward gaining a real understanding of water in food systems, incorporating the view of water as a reactant and a "catalyst", water as a structure influencing material, and water mobility as a function of external and internal influencing factors.

It also was focused on the conversion of extrusion-cooking technology into extrusion-cooking science, considering the mechanical aspects of the extruder as well as the chemistry and kinetics of the extruder as a pressure-shear-temperature reactor.

Another component of the studies was dedicated to the development of new in-line sensor concepts for real-time measurement encountered in a variety of food processing environments, through the synthesis of new chemical compounds and new materials which may serve as sensors and the development of new systems which provide measurable responses leading to improved process efficiency and product quality through automation and control.

4.B. Summary of the Most Important Results

4.B.1. Water Phenomena Project (Formerly The Water Relations Project)

4.B.1.1. Migration and Material Changes

Our studies of isothermal moisture transfer in starch systems have led to the development of a simple formula to represent diffusivity as a function of moisture concentration.

In non-isothermal studies of triply layered starch-sucrose-starch, it was found that sample size affects the stress profiles. In smaller samples, tensional stresses were found in the outer layers; in larger samples, compressional stresses were found in the inside layers. In the drying of concentric, viscoelastic cubes of starch-sucrose-starch, moisture transfer was initially *from* the core and later, *to* the core due to mass transfer potentials in addition to diffusional transfer. Analytic solutions may be adaptable to a PC with sufficient computational capability although they will need to be based on restrictive assumptions.

Responding to the need to simplify methods for predicting hygrostress formation during transient state mass transfer processes without use of main frame or super computers, we have developed analytical solutions which are adaptable to PC or programmable hand calculators.

Parametric studies continued to provide information useful for process optimization based upon heat transfer, moisture transfer, hygrostress formation and crack propagation.

4.B.1.2. Diffusion and Molecular Structure

Computer modeling of protein structure using thirty of the best resolved crystallographic structures as a data base was accomplished. When algorithms are run over the entire protein, irrespective of helix or strand boundaries, many helices and strands appear longer by several residues than the crystallographic data show. In the early stages of folding, up to and including the molten globule state, secondary structures are actually longer than the native form. This analysis of static structures may lead to insight into kinetic events.

Glass transition behavior research has provided a state diagram for sucrose which allows the prediction of the physical state as a function of temperature and concentration. A maximally freeze-concentrated sucrose matrix contains about 80% sucrose showing a glass transition (T_g) at -46°C and an onset temperature of ice melting at $T_m = -34^\circ\text{C}$. In concentrated solutions, ice formation is kinetically controlled by viscosity of the solution. Maximum freeze concentration can be achieved at a temperature above T_g but below T_m . The T_g for sucrose decreases with increasing water content and can be predicted using a linear relationship between water activity (a_w) and T_g or using the Gordon-Taylor equation which allows calculation of T_g values from weight fractions.

Diffusion of volatile compounds in amorphous sucrose, above T_g , follows the WLF equation. In this study, crystallization occurred simultaneously with diffusion. Studies of the influence of hydration on the physical properties of lysozyme have used luminescent signals from intrinsic tryptophans to probe the interior of the protein and extrinsic covalent probes attached to the imidazole side chain of the unique histidine (HIS-15). The tryptophan studies indicate an increase in protein flexibility over the range of humidities from 0-90%. Both fluorescence and EPR probes attached to HIS-15 indicate that even at high RH values (around 90%) the bound water does not change the surface polarity of dry protein powders. This finding supports non-uniform distribution of water.

We have established the solution conditions for our model proteins (Bovine Alpha Lactalbumin and Cytochrome C from Tuna) and can produce conformationally stable proteins in native, molten globule and unfolded states. This establishes the protocol for subsequent efforts.

To utilize our previously reported volume change of aliphatic groups in transfer from water to alcohol ($26.2\mu\text{l}/\text{A}^2$ Mole) data requires solvent accessible surface area of native protein. We have now achieved this via computer modelling.

We have developed and placed in operation a flow system which allows us to vary the hydration level of lysozyme samples in situ in the spectrophotometer. We are now able to measure and compare luminescence intensities for the same sample at different hydration levels. We have shown the phosphorescence and fluorescence intensity of lysozyme are sensitive to hydration level, therefore, serving as intrinsic surface hydration probes. The generality of these phenomena to other proteins and the influence of carbohydrates and lipids is being investigated.

Evaluation of the suitability of carbohydrate mixtures on oil encapsulation as a function of time, temperature and moisture content has indicated that collapse of the matrix led to partial release of the oil, while in crystalline systems, all oil was released.

4.B.1.3. Reaction Chemistry

Studies on the quantification of lysozyme glycosylation and aggregation have shown that both occur even at room temperature. At 65% RH and 50°C , monomers and aggregates were observed after 15 days. At room temperature fewer aggregates were found after 15 days, but these increased after 4 months. These room temperature findings bear directly on product quality changes during storage, such as in powdered milk.

Work has been completed on the semi-quantitative analysis of flavor generation from a series of samples by purge-and-trap gas chromatography-mass spectrometry. Heated native lysozyme generated important flavor compounds such as sulfur dioxide, dimethyl disulfide, dimethyl trisulfide and indole. These results suggest the oxidation of methionine and tryptophan.

Volatile analysis has been found to be a more sensitive indicator of glycation than previous HPLC peptide mapping and FAB mass spectrometry. The formation of alkylpyrazines, the most important flavor compounds in roasted and baked foods, has led to the hypothesis that asparagine or glutamine residues are deamidated.

A sensitive enzymic method to measure ammonia formation resulting from deamidation of asparaginyl and glutamyl residues of lysozyme has been developed. It was used to verify that deamidation is a major source of formation of nitrogen compounds, such as pyrazines.

4.B.2. Extrusion Cooking Technology and Science

4.B.2.1. Engineering Analysis of Extrusion

In numerical simulation of transport, the emphasis has been to investigate the shear and temperature effects on conversion of starch using chemical kinetics and rheological data available for the starch. It is confirmed that shear effects dominate over much of the operating range of the extruder. At high temperatures, the thermal effects are dominant. The numerical results on specific mechanical energy (SME), conversion, heat transfer, etc., have been presented as correlating equations, which will be valuable in scale-up and design of operation of the extrusion process.

The project continued to make progress in engineering analysis of single and twin-screw extruders as well as in defining the chemical changes in cereal flours which affect extrudate quality. In engineering studies, the transport phenomena in twin screw extruders, for both tangential and self-wiping screw geometries, have been successfully simulated to calculate pressure, temperature and velocity fields, mixing characteristics, and shear rate variation inside the extruder. The results have been compared with those for single screw extruders, indicating an improved mixing and very similar pressure and temperature increases in twin screw extruders. The results also agree qualitatively with the experimental observations. The residence time distribution (RTD) prediction from the finite element model (FEM) simulation of a single-screw extruder has been improved by incorporating variable barrel temperatures into the model. Results show that the variable barrel temperature has little effect on the RTD, and that a significant spread in the residence time function, $f(t)$, occurs as the channel length is increased. The results provide design guidelines for the selection of single vs. twin-extruders and to appropriately configure an extruder.

The experimental work focused on quantifying the response time of the traversing thermocouples developed for the single screw extruder. Correlations between the response time and the heat transfer rate will be obtained with a simple heat apparatus which is already built. An Anemometer and Infrared Imaging system were acquired and are being integrated into the ZSK-30 twin-screw extruder. Also, elapsed time devices were installed to measure the heater energy input.

In experimental studies on the ZSK-30 twin-screw extruder, experiments were carried out to measure heat transfer rate and the heat transfer coefficient between the extrudate and the barrel for corn meal and Hylon VII starch at two different moisture contents (22% and 30%). A wide range of heat transfer coefficients ($100-3000 \text{ w/m}^2\text{K}$) was obtained. In a series of trials known as the group experiments, initial trial runs were made with hard (14% protein) and soft (8% protein) wheat flours to obtain a window of operating conditions suitable for the ZSK-30 which were: 16%-20% moisture content, 300-500 rpm screw speed, 100-400 g/min throughput

and 160-190°C die temperature. This would serve as a guideline in design of extrusion experiments. It was also shown that addition of a reverse screw element (1% of total length) at two different moisture levels (18% and 29%) and at two different screw speeds (200 and 400 rpm), increased the SME but did not increase the residence time. Correspondingly, the springiness and bulk density of the extrudate decreased as a result of the addition of a reverse screw element.

4.B.2.2. Measurement of Reaction Kinetics and Physical Properties Relevant to Extrusion

The glass transition temperature (T_g) decreased with an increase in the SME. Also, the melting temperature of Amioca starch was found to be highly influenced by the storage temperature above 35°C.

In GEL and MELT studies it was found the Gel and Melt starches are different in molecular size (after DMSO treatment), water sorption isotherm, solubility and retrogradation as well as iodine staining capacity, birefringence pattern and shape of starch granules. The molecular size of Gel is larger than that of Melt. The hygroscopicity and solubility of Gel are lower than that of Melt. Gel retrogrades but Melt does not.

It was found that below a certain value of moisture content and degree of conversion, the starch particles were not connected to each other. This study is aimed at understanding the transition from powdery state to continuous fluid state which must be taken into account in the modeling of extrusion.

In physical properties measurement, the glass transition temperatures (T_g) of zein, gliadin and glutenin at various moisture contents were determined. It was found that T_g of zein and gliadin were considerably lower (15-30°C) than that of glutenin and wheat gluten. Gliadin with 24% and 30% moisture contents showed another transition between 90-110°C. The temperature dependence of the apparent viscosity of glutenin at 40% moisture was found to follow the Arrhenius equation. The glass transition temperature (T_g) of corn flour extrudates decreased with an increase in SME, moisture content and amylose/amylopectin ratio. Correspondingly the crispness of extrudates decreased and denseness increased.

The thermal conductivity of granular starch increased with an increase in mechanical pressure, through changes in the porosity. Thus, during extrusion, the thermal conductivity of starch will change along the length of the extruder which will affect temperature-time history of the extrudate.

4.B.2.3. Chemical Changes During Extrusion

Phenolic acids were found to enhance increases in fluorescence anisotropy and molecular weight during extrusion, but only in the presence of atmospheric oxygen or an added oxidizer. Comparative extractions of phenolics from corn meal and extrudates demonstrated that the fraction of phenolics bound to starch increases during extrusion; also, zein undergoes complexation with starch, with the involvement of phenolic acids. Correlations of several appearance, taste, and texture qualities of extrudates were obtained as functions of their fluorescence anisotropies. This revealed that all these properties monotonically increase or decrease with increasing anisotropy, but that the changes occur abruptly at some critical anisotropy value, that may correspond to a critical structural change, such as a phase change.

During extrusion, the presence of ferulic acid and of smaller amounts of other phenolic acids in corn meal was confirmed by thin-layer chromatography. A high-molecular weight complex of ferulic acid with protein and/or carbohydrate was found to be much greater in extrudates than in uncooked corn meal. Its amount and molecular weight increased with increas-

ing extent of conversion. Model extrusion experiments showed that addition of ferulic acid produced greater cohesiveness and higher anisotropy. The generation of free radicals in extrudates, as measured by electron spin resonance (ESR) was shown to depend not only on mechanical energy input, but also on temperature, moisture, and starch type. Higher ESR signals were obtained for sample containing high amylose.

In the study on structural transformations of carbohydrate and protein during extrusion, a new strategy was devised for the direct solid-state assay of buried and exposed -SH groups and -S-S bonds in corn meal-based materials. This assay was highly reproducible and agreed with direct amino acid analysis. Following extrusion, a small amount (4-6%) of the -S-S bonds were converted into -SH groups in some, but not all, samples. These results show that small, but not extensive, changes in the ratio of -SH to -S-S groups occur during extrusion.

Gel filtration chromatography with application of the universal calibration concept, was applied to quantify extrusion-induced starch fragmentation in Pure N Thick corn flour. Significant fragmentation was seen to occur in each of the extrudates, as evidenced by the marked shift of molecular weight profiles towards lower molecular weight ranges. Weight average molecular weights decreased from 18,800,000 in native "Pure N Thick" to a range of 12,000,000 - 5,950,000 in extrudates. In extrudates, lipid yields were found to be low, indicating that lipids in the extruded corn meal products are highly complexed and/or strongly occluded in the starch/protein matrices. An inverse relationship between lipid extractability and lipid oxidation was obtained. Increased residence times resulted in increased carbonyl contents.

A model system containing corn meal blended with 60% casein was employed for the study of bonding interactions between biopolymers during extrusion. It was found that after extrusion, the total extractable free amino acids with soluble peptides increased by a range of 37.7% to 101.6%. Total amino acids with soluble peptides increased by two- to three-fold after extrusion mainly due to the presence of the small peptides suggesting the protein fragmentation or hydrolysis during the extrusion processing. An attempt was made to monitor the extent of deamidation of proteins in high gluten wheat flour after undergoing particular heat treatments. It was found that the extruded sample gave the highest degree of protein deamidation, the boiled sample gave the second highest, and the cooked sample gave the lowest deamidation value.

A study of the chemical interactions of lipids as they pertain to amylose and amylopectin ratios in extruded corn flours showed that amylose is more effective at complexing with lipid during extrusion. However, it was found that in both high amylose and high amylopectin extrudates the majority of the lipid was released with water saturated butanol extraction. This indicated that the majority of the lipid in these extruded systems is present in some close molecular association with the starch fraction.

Extrudate samples were analyzed for the volatile compound composition. The volatile compounds were identified by GC-MS and included furanones, phenols, aldehydes and pyrazines. Over 100 compounds were identified in the condensates collected at the extruder die. The compounds identified indicated that oxidation, hydrolysis, thermal degradation, decarboxylation and Maillard reactions occurred during extrusion.

The effect of gluten on the retention of model flavor compounds during extrusion was evaluated. With the increased content of gluten, more limonene is recovered by extraction before and after extrusion. Treatment with dithiothreitol was found to increase the recovery of limonene after extrusion. Treatment with enzyme increased recovery of limonene both after and before extrusion.

4.B.3. In-Line Sensors Concepts

4.B.3.1. On-Line Slit Rheometer

The on-line slit rheometer is now available for testing. Some improvements include a control loop which is ready to be tested on different systems. On-Line viscosity during oat-mix extrusion was measured. Experiments with polymers and non-cooking doughs measuring entrance pressure drop in the modified slit rheometer were performed with very good results. In addition, a screw pump rheometer for studies with cooking and non-cooking food systems was developed. Fitting is underway to allow simultaneous viscosity measurements with the on-line slit rheometer.

4.B.3.2. Ultrasonic Sensor

The potential of an on-line ultrasonic rheological sensor using PDMS (polydimethylsiloxane) a well characterized non-food system, has been established. The optimum experimental set-up and procedure have been identified and implemented to study ultrasonic wave propagation through the viscoelastic material. A transducer holder was fabricated to maintain contact pressure and the sample cell was modified to minimize dimensional effect. Output signal processing was improved to treat the raw data. The sampled data points were increased from 256 to 4096. A series of dough samples with 5% difference in moisture were analyzed. Correlations are underway.

4.B.3.3. Thermomoisture Sensor

Subsequent to the development of an actual sensor, a computer program designed for sensor response simulation, under non-ideal conditions, has been experimentally validated. The influence of non-uniform moisture distribution, non-uniform temperature distribution, and temperature dependent heat transfer properties on sensor response has been studied. A test cell for use with powder materials has been designed. Testing of the cell has been initiated.

This sensor has been tested with several member companies. A patent application is underway.

4.B.3.4. Porous Glass Fiber Optic Sensor

A ruggedized sensor has been designed, fabricated, and successfully tested in CAFT's EDC Hot Air Impingement oven for qualitative response. Discussions have been initiated with a member company to assist in developing a calibration chamber for the sensor. Basic optoelectronic components for the sensor system have been identified. Sensor system prototype design has been initiated.

4.B.3.5. Automated Fiber Optic Moisture Sensing System

Components and coupling design to interface the prototype optical source and detection system previously demonstrated, were identified. The focus is on demonstration with the CAFT fluorescent film sensor. A blue laserdiode has been identified for application in the fluorescence region. Also a UV transmitting fiber with low attenuation <30 dB/Km at .4 μm was identified. An asymmetric bidirectional coupler has been designed for enhanced crosstalk isolation with

multiple sensors. Combination of components into the overall remote data acquisition and analysis system is in progress.

4.B.3.6. Fluorescent Films for Fiber Optic Moisture Sensing

The utility of poly(vinylpyrrolidone) (PVP) polymeric thin film impregnated with an aluminum-morin compound for visible wavelength detection of moisture in a specific, reversible and rapid manner was further investigated. A theoretical model has been developed to evaluate experimental results and to provide a quantitative framework for the optimization of sensor performance.

The results from the fiber optic based system, mounted on the optical bench, have clearly shown that it is possible to correlate fluorescence signals as a function of relative humidities. The effect of modifying the film thickness was studied and related to the sensor response time. Temperature effects on the sensor response were investigated. Current results show that active thin films (20-50 μm) can be easily deposited on glass substrates and that good sensitivity is observed to at least 82°C.

Thermal analysis of the film has shown that the aluminum/morin complex is stable up to 190°C; at this point a phase transition of the PVP is observed.

A high temperature prototype design has been initiated.

4.B.3.7. Ceramic Sensing Materials

Two types of moisture sensors, impedance-type $\text{Na}_3\text{HGe}_7\text{O}_{16} \cdot x\text{H}_2\text{O}$ and emf-type NASICON-based electrolyte were developed based on the structural properties and previous testing of materials. Both humidity sensing materials were tested for reproducibility of moisture sensing by measurements at different moisture levels with cycling for long periods of time and good results were obtained.

The effects of other gases ($\text{C}_2\text{H}_5\text{OH}$, $\text{C}_2\text{H}_3\text{O}_2$, $\text{C}_2\text{H}_4\text{O}$ and NH_3) on the moisture sensing properties and stability of materials were evaluated on both of the above materials by emf and AC-impedance respectively. Both sensors show good reproducibility and durability.

4.B.3.8. Piezoelectric Nylon Moisture System

The data base for temperature-stabilized nylon 11 polarized films has been essentially completed. These materials show excellent sensitivity and response to moisture changes. Collection of the data for temperature-stabilized nylon 7 polarized films is underway.

Moisture sensing results of a new piezoelectric material consisting of coprocessed "bim laminate" of nylon 11 and poly(vinylidene) fluoride (PVF₂) suggest that the sensitivity of this new material is more than an order of magnitude greater than that of nylon 11 alone.

A sheet of nylon sensor material was prepared and 3 sensor elements were made. One element was kept for a control, the other two elements were sent to member companies and exposed to food processing environments, after which the moisture responses of the three elements were compared. No change in performance characteristics was seen in the exposed elements.

4.B.3.9. Electro-Optical Inorganic Film Moisture Sensor

An electrical sensor based on an interdigitated gold electrode and several polymer films including Lithium Nafion and Lithium Sulfonate was designed. Lithium Sulfonated PEEK with different degrees of sulfonation was synthesized in order to investigate film properties and response time of the electrical measurement. The electrical sensor range of moisture response up to 180°C was tested, the low moisture range is very responsive. Model complexes of Co(III), Ni(I) and Cu(II) with 1, 4, 7 trimethyltri-amino cyclononane were synthesized and characterized. Their response to water in solution and when immobilized on films was shown. Initial experiments to incorporate the metal complexed in Zeolite Y are underway.

A prototype for the electrical sensor is being constructed and will be capable of field testing once fabricated.

4.C. List of All Publications

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4.D. List of All Participating Scientific Personnel

<u>Name</u>	<u>Degree Earned While on Project</u>
Abib, A.H.	Ph.D. Degree
Adachi, J.D.	
Alaimo, L.H.	
Avramidis, K.S.	
Bassompierre, M.	
Bhakuni, Sudhir	Ph.D. Degree
Breslauer, K.	
Buera, M.P.	
Carillo, P.J.	
Chan, K.Y.	M.S. Degree
Chang, C.N.	Ph.D. Degree
Chang, K.L.B.	Ph.D. Degree
Chatterjee, S.	Ph.D. Degree
Chedid, L.L.	
Chen, J.	
Chiang, W. C.	
Cho, M.	
Chung, T.Y.	Ph.D. Degree
Cocero, A.M.	
Daun, H.	
Dervisoglu, M.	
Ding, J.	
Drake, R.R.	
Dus, S.J.	
Esseghir, M.	
Feng, S.	
Frenkel, C.	
Frost, D.J.	
Gernstern, B.	
Giannakakos, P.	Ph.D. Degree
Gibson, S.M.	
Gilbert, S.G.	
Goedeken, G.L.	
Gopalakrishna, S.	
Govindarajan, S.	
Greenblatt, M.	
Gross, K.S.	
Gupta, M.	Ph.D. Degree
Guzman, L.	
Halek, G.W.	
Harriman, R.W.	

Hartman, T.G.
 Hartwick, R.
 Hayakawa, K.
 Henrikson, F.W.
 Herskowitz, G.
 Ho, C-T.
 Hwang, S.H.
 Il, B.
 Isied, S.S.
 Itaya, Y.
 Izzo, H.V.M.S. Degree
 Jaluria, Y.
 Jiang, T.S.
 Kahn, P.C.
 Kaletunc, G.
 Kang, B.H.
 Karathanos, V. T.
 Karel, M.
 Karmas, R.
 Karwe, M.V.
 Kim, I. H.
 Kodama, T.
 Kokini, J.L.
 Kontominas, M.
 Kwon, T.H.
 Lai, L.S.Ph.D. Degree
 Lee, H.O.
 Lee, J.W.
 Leslie, R.B.
 Levi, G.
 Lints, R.R.
 Liu, J.N.
 Luan, H.M.S. Degree
 Ludescher, R.D.
 Ma, S.
 Madeka, H.
 Mezhoudi, M.
 Mitcham, E.J.
 Newman, B.A.
 Ogawa, M. Y.
 Oh, Y-C.
 Pandya, R.N.....Ph.D. Degree
 Papantonis, N.C.M.S. Degree
 Pedersen, H.
 Petrone, L.
 Potenza, J.A.
 Ramon, O.
 Read, S. M.

Rodis, P.
 Roos, Y.
 Rosen, J.D.
 Saggese, S..... Ph.D. Degree
 Saguy, I.
 Sakai, N.
 Saravacos, G.
 Sastrohartono, T. Ph.D. Degree
 Scheinbeim, J.I.
 Schugar, H. J.
 Serafin, F.L.
 Sernas, V.
 Shah, N.K.
 Shahriari, M.R.
 Shimada, Y.
 Shu, C-K.
 Sigel, G.H.
 Smith, T.
 Solberg, M.
 Song, Y.
 Strauss, G.
 Szu, S.P.
 Takacs, S.F.
 Takese, Y.
 Tanase, S.
 Timpa, J.D.
 Tong, C.H.
 Tsai, M.
 Tsukada, T.
 Uq, D..... Ph.D. Degree
 Waks, M.
 Wang, E.
 Wang, J..... Ph.D. Degree
 Wang, W.
 Wasserman, B.P.
 Wen, L.F. Ph.D. Degree
 Wishart, J.F.
 Wu, A. Ph.D. Degree
 Yam, K.L.
 Ybe, A. Ph.D. Degree
 Yilmazer, G.
 Zhao, B.
 Zheng, X.
 Zhang, Y.

5. Report of Inventions

No patents issued.

6. Bibliography

The Following is a list of publications generated in the Basic Research Program to which this agreement (DAAL03-K-0174) is contributing.

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